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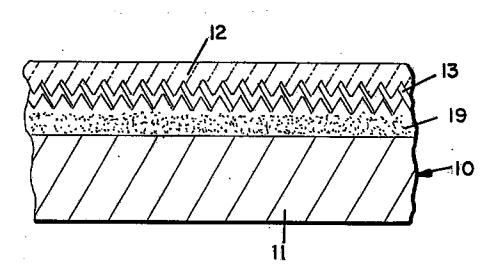
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(54) Title: METHOD OF ENHANCING THE VISIBILITY OF DIFFRACTION PATTERN SURFACE EMBOSSMENT



(57) Abstract

Methods for enhancing the visibility of and imparting a metallic luster to holographic and diffraction pattern surface embossments on transparent media such as plastic films (12) are disclosed. Enhancement is achieved by (1), metallic effect dyes or (2), metal flake inks containing minimal quantities of resinous binders (3), inks containing crystalline pigments that have metallic visual effects, or (4), inks containing pigments formed of optical stacks. The solvent used during application of the dye or ink must not be a solvent for the embossed surface.

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# METHOD OF ENHANCING THE VISIBILITY OF DIFFRACTION PATTERN SURFACE EMBOSSMENT

### 1. Field of the Invention

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This invention relates to the decoration of sheeting and, more particularly, to the decoration of materials such as thin transparent plastic films. This invention also relates to the embossment of plastic film and more particularly to improved methods for making embossed patterns visible to the human eye.

### 2. Description of the Prior Art

There are wide-spread applications for embossed plastic films. One example is provided by plastic films embossed with holographic patterns. These are used in application where security sensitive items such as holographic labels are used. They are also used for decorative applications such as self adhesive stickers and gift wrap.

The above-described embossments on plastic films are usually made clearly visible by depositing a thin layer of metal (usually aluminum) on the embossed surfaces to give a mirror-like luster which intensifies the visibility of the embossments. This thin layer of metal is generally applied to the surface by a process known as vacuum metallization. In that process, the film to be metallized is placed in a large vacuum chamber and, after a high vacuum is achieved, is passed over a series of crucibles or evaporation boats containing molten metal. The metal deposits from the vapor phase onto the moving plastic web. This method of metallization can be conducted at high speeds (up to 2000 ft. per min.) on wide webs, but has several significant disadvantages. One disadvantage is that the metallizing equipment is very expensive. A second disadvantage is that the metal can only be applied as a uniform coating or, by using appropriate masks, in stripes running in the machine (long) direction of the plastic film web. If it is desired to create a discreet metallized pattern, this is usually accomplished by first metallizing the surface overall, there etching away the unwanted metal by use of a corrosive etchant such as caustic soda The etching process is expensive because it entails an additional or an acid.

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manufacturing step, specialized machinery, and handling and disposal problems with the etchant. Another disadvantage of the vacuum process is that the metal can only be applied at one level of deposition, whereas it is often desirable to have different levels of metal deposited at different points on the plastic film to produce partial transparency or other special effects. Still another disadvantage of the vacuum metallization technique is that it is not suited for use on narrow webs or small rolls of film, and is especially uneconomical to use when the film is in sheet form.

It is accordingly an object of the present invention to provide improved and simplified methods to render images visible.

Another object of the invention is to allow the application of metal or metallic effect materials to embossed films without the need for expensive vacuum equipment.

It is yet another object of the invention to allow the metallic decoration of embossed films on conventional printing presses.

A further objective is to provide methods for producing metal or metal-like deposits on discreet areas of embossed films without the necessity of using corrosive etchants.

An additional object of the invention is to provide means to obtain a wide variation of metallic or metal-like deposits ranging from almost completely transparent to totally opaque, all produced in-line on a single standard press.

A still further object of the invention is to allow the metallization of small quantities of embossed film in an economical manner.

### SUMMARY OF THE INVENTION

In accomplishing the foregoing and related objects, the invention involves the printing of a metal flake ink or "metallic effect" ink onto a transparent or translucent film which has an embossed pattern on at least one surface. The pattern may be of a spacial scale suitable for a diffraction pattern, a holographic image, or a microscopically-visible image. It might also be of a larger spacial scale suitable for macroscopically visible images or patterns. The "metallic effect" ink or metal flake

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ink contains a solvent and/or diluent liquid which may be water or an organic liquid, but which is not a significant solvent for the plastic film or the embossment.

The systems of the present invention, which systems substitute for vacuum-deposited metal films, fall into four types. The type one system involves printing inks which employ, as their pigment, a special form of metal flake. This form is highly reflective, very thin (preferably 100 to 500 angstroms), and large in length and width (diameter) (functionally 0.1 to 50, and preferable 10 to 50 microns). Of particular preference is a new class of such flakes which are themselves embossed.

The type two systems involve the application of certain dyes, which, when properly applied, simulate and enhance the visual effect that would be brought about by the vacuum-deposited metal films.

The type three systems involve inks which use, as pigment, a crystalline material such as graphite or sulfides or selenides, including MoS<sub>2</sub>, PbS, FeS<sub>2</sub>, CoS, Ni<sub>2</sub>S<sub>2</sub>, SnS<sub>2</sub>, ZrS<sub>2</sub>, possessing delocalized electrons.

The type four systems are inks which use, as pigment, particles of optical stacks, particularly those with an embossed surface.

In a typical application, a clear sheet of plastic would be embossed on one side. The embossed side would then be printed with the inks involved in this invention. The printing may be selective, that is, some of the sheet may be rendered opaque, some rendered translucent, some rendered transparent, but colored, and some not printed and therefore transparent. The embossed and printed side may then be bonded to a printed side of an opaque card of cardboard or plastic in order to complicate forgery or alteration of the card.

### BRIEF DESCRIPTION OF THE DRAWINGS

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The character of the invention, however, may best be understood by reference to one of its structural forms, as illustrated by the accompanying drawings, in which:

Fig. 1 is a cross-sectional view of a laminated decorative sheet embodying the principles of the present invention,

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Fig. 2 is a cross-sectional view of a second laminated sheet embodying the principles of the present invention,

Fig. 3 is a flow chart expressing the process steps embodying the principles of the present invention, and

Fig. 4 is a schematic representation of a process for creating laminated decorative sheets embodying the principles of the present invention.

### DESCRIPTION OF THE PREFERRED EMBODIMENT

Referring first to FIG. 1, wherein are best shown the general features of a laminated decorative sheet embodying the principles of the present invention, the sheet, denominated generally by the numeral 10, is shown to include a backing sheet 11, and a transparent cover sheet 12 adhesively bonded to one surface of the backing sheet 11 by adhesive 19.

The inside surface of the cover sheet 12 is embossed with a diffraction grating or holographic image. This inside surface is printed with a layer of the type one, two or three ink 13, which simulates the optical effect of metallizing that surface, when the cover sheet 12 is viewed from its outer surface. In this simple laminate, the optical effect not only provides a decorative aspect, but also is difficult to duplicate by forgers.

A more complex structure is presented in Fig. 2. In this structure, the inside surface of the backing sheet 21 is printed with information 24 which requires protection from forgery or alteration. The transparent, embossed cover sheet 22 is printed with the layer of type one, two or three ink 23, but not uniformly. In zone 25 and 26, the ink is thick and essentially opaque. The printing 24 does not show through the cover sheet 22. In zone 27, the ink is thin and translucent so that the printing 24 is visible but somewhat obscured. In zone 28, there is no ink so the printing is clearly visible through the adhesive 29 and the embossing is invisible. This structure is very difficult and complex to duplicate or forge. It therefore provides an effective anti-forgery and anti-alteration system. Because the ink is applied using a

printing process, precise control of the zone or windows in the sheet and the pattern of decoration or obscurity can be achieved.

Fig. 3 provides a flow chart of the steps employed in a typical application of the present invention. In step 1, the card or backing sheet is formed. In step 2, the card in printed with the sensitive information. In step 3, the cover sheet is embossed. In step 4, the embossed surface is selectively printed with the type one, two or three ink. In step 5, the printed side of the card and the printed side of the cover are adhesively bonded to form the product.

In an alternative, the sensitive information could be printed on the embossed surface before or after the ink of the present invention is printed on the embossed surface.

Fig. 4 presents a schematic representation of a process by which the product embodying the present invention might be constructed. Supply roll 51 provides an elongated belt of backing sheet 52. As the backing sheet 52 passes along the path, printing roll 53 prints sensitive information 54 on the surface of the backing sheet 52. Simultaneously, feed roll 55 provides smooth cover sheet 56 to softener lamp 57, embossing roll 58, and backing roll 59. The inside surface of the cover sheet is embossed.

The embossed surface 61 is then selectively printed with the metallic-effect ink 62 by printing head 64. The adhesive 66 is the applied to the cover sheet by adhesive head 68. Then roll 69 presses the backing sheet 52 and the cover sheet 56 together to adhesively bond the printed surfaces. The resulting laminate is stored on take-up roll 71. The product is subsequently cut into useful sizes.

The systems of the present invention, which systems substitute for vacuum-deposited metal films, fall into four types. The type one system involves printing inks which employ, as their pigment, a special form of metal flake. This form is highly reflective, very thin (preferably 100 to 500 angstroms when aluminum is employed), and large in length and width (diameter) (functionally 0.1 to 50 and preferable 10 to 50 microns when aluminum is employed). Of particular preference are a new class of such flakes which are themselves embossed.

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The metal flakes are used in two separate diameter ranges and thereby function in two separate regimes. The first preferred regime involves flakes with average diameters greater than or significantly greater than the average peak-to-peak distance (wave length) of the embossment. These flakes appear to bridge across the embossment peaks to achieve their mirror-like optical effect.

The second regime involves flakes with diameters smaller than the wave length of the embossments and especially smaller than the slope (peak-to-valley) length. These flakes tend to leaf the slopes of the embossment to achieve their mirror-like optical effect.

The type two systems involve the application of certain compounds, which, when properly applied, simulate and enhance the visual effect that would be brought about by the vacuum-deposited metal films.

The type three systems involve inks which use pigments such as graphite, which are strong optical absorbers.

The type four system involves inks which use, as pigment, particles of optical stacks, and particularly those with an embossed surface.

The metal flakes of the invention are preferably those produced by vacuum metallizing a solvent-soluble coating on a supporting substrate to produce a mirror-like deposit generally between about 100 to 500 Angstrom units (for aluminum) in thickness. The substrate and its coatings are then immersed in a solvent bath to dissolve the soluble coating so that the metal layer floats free in the solvent. After repeated rinsings and filtrations or centrifugations, the dispersion containing the metal layer becomes nearly free of the solvent soluble coating material. The puritied metal layer is then broken up into fine particles by high shear mixing. The resulting material is finally diluted with an appropriate solvent and a vehicle or binder is added to form the final ink. It is critical for the present invention that the binder-to-metal flake ratio be carefully controlled as will be discussed later. Such inks, when properly prepared, are of low viscosity. They can be applied by spray or printed by gravure of flexographic means onto smooth transparent films. This produces a mirror-like metal surface when viewed through the unprinted surface of the

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transparent film (known as second surface viewing). A representative metal flake product of the aforementioned type is known by the trademark Metalure<sup>TM</sup>, produced by the Avery Dennison Corporation.

The "metallic effect" dyes of the invention are preferably selected from the group consisting of crystal violet and rhodamine B. Other dyes that also function are fuchsin, methyl violet, safranine, fluorescein, rhodamine, and mauveine. Type three crystalline materials having delocalized electrons also function for this application. Although the preferred material is graphite, other appropriate materials are FeS<sub>2</sub>, MoS<sub>2</sub>, or PbS. Appropriate materials must (1) have a crystalline structure with delocalized electrons, (2) have high optical absorption, greater than 10<sup>6</sup>cm<sup>-1</sup>, with good reflectivity near the surface, and (3) have a smooth surface on a microscopic scale. Furthermore, because the preferred method of attaching the crystalline material to the embossed surface involves application as pigment in a binder, it is desirable that the material be capable of achieving high pigment/binder ratio. Other possible materials are potassium permanganate, iodine, cadmium selenide, polydiacetylenes, silicon or germanium.

In the practice of the invention, a plastic film is embossed using well known methods (e.g., embossing the surface of a polyethylene teraphthalate film by pressing it in contact with a heated nickel embossing shim at high pressure). Such embossments may have a depth of less than 0.5 microns, and a spatial frequency (wave length) of 1.5 microns or less. After the transparent film is embossed, the pattern is discernable if viewed under favorable lighting condition, such as bright illumination by a point lighting source, but very faint or virtually invisible if viewed under more diffuse (e.g., fluorescent) lighting conditions. The pattern on the film is made more strongly visible by being printed with a metal flake ink containing not more than about 2.5 parts of binder solids (by volume) per part of metallic flakes. Above 2.5 parts of binder per part of metal pigment, the embossed pattern loses contrast because the binder fills in the embossed pattern and "blinds" or "quenches" it. This is because the binder has a refractive index which is close to that of the film so that, when it fills in the embossment, the appearance becomes that of a flat,

unembossed film. If the binder content drops below about 0.1 per part of metallic flakes, the flakes are not adhered together tenaciously, and may be removed easily, for example, with adhesive tape.

More specifically, the functional range of binder to flake ratio is 0.01 to 2.5 and the most preferred range is 0.5 to 1.0. Binders include vinyl chloride/vinyl acetate copolymers, vinyl, acrylic, urethane, polyester, terpene/phenol, polyolefin, silicone, cellulosic, polyamide, and rosin ester resins. Polyamide and acrylic provide the best adhesion to Mylar.

Although the preferred method of attaching the flakes to the embossed surface would be with and by a binder, other methods are possible. For example, a short wash of the embossed surface with a solvent to the surface, sufficient to make the surface tacky, could then be followed by exposure of the flakes to the still tacky surface to make them stick to the surface. Similarly, hot air, or radiant heat or warm flakes could render the surface tacky for application and holding of the flakes. The simplest approach would be to apply the flakes to the surface while it is still soft and tacky from the embossing step. It would also be possible to apply a holding sheet over the back of the flakes on the surface to hold the flakes to the surface. The flakes have a tendency, even without binder, to stick to the embossed surface and to each other, weakly, but sometimes usefully.

A variation of this basic concept of metal-flake based ink, which variation appears to be novel itself involves causing the flakes themselves to have embossed surfaces. This process is described in detail in a copending application. To accomplish this, the above-described metal flake forming process is modified by forming the original metal sheets on a soluble substrate which is itself embossed. This single modification of the conventional process causes the resulting metal flakes to be embossed on both sides. When these flakes are used to form ink and then printed on embossed, or even on smooth plastic film, a dramatic visual effect results.

Alternatively, the pattern on the film is made more strongly visible by the application of crystal violet dye or rhodamine dye, from solution, to obtain final dried coating weights of dye between a minimum of 10<sup>-3</sup> grams per sq. meter and maximum

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of 1.0 grams per square meter (gsm). Below the minimum coating of dye, the embossment receives a strong purple coloration, but no strengthening of the embossing-pattern visual effect occurs. At the lower application weights of the dyes, the metallic effect can be enhanced if the dye is backed by a black coating or a metallic coating (e.g., by conventional aluminum inks). The unusual interaction between dyes, in sufficient thickness, and the embossed plastic surface simulates the optical effect of the metallized embossed surface.

Another method to make the pattern on the film more strongly visible is to apply a coating using pigments having strongly delocalized electrons such as graphite and sulfides.

A particular type of pigment which would have application in the inks of the present invention is called an optical stack and is described in U.S. Patent No. 4,168,986 to Joseph J. Venis, entitled, "Method for Preparing Lamellar Pigments", and U.S. Patent 5,084,351 to Roger W. Philips et al., entitled, "Optically Variable Multilayer Thin Film Interference Stack on Flexible Insoluble Web". The teachings of both of these patents are incorporated herein by reference.

By precisely controlling the thickness of layers of optically-effective materials and the formation of optically-effective layer boundaries, interesting and useful optical effects can be produced when light impinges on the optical stack.

Of particular interest for this invention would be a process of making optical stack pigment flakes that are initially formed on an embossed carrier (or embossed carrier-release layer). In that way, the optical stack would carry the embossment. The stack could be formed on the embossed carrier either with the front surface against, or away from the carrier. Furthermore, the layer adjacent the carrier may be applied to fill the embossment so that subsequent layers have parallel boundaries

and/or the first layer may be a supplemental cover layer which simply places an embossed cover on the rest of the stack. Alternatively, the stack layers can be laid on so that each follows the embossment or proportionally follows the embossment (i.e., uniform frequency but reduced amplitude). When the stack is removed from the carrier and employed as a pigment, the optical effects of the stack and embossment on each stack particle would occur. This invention would also include forming the optical stack on an embossed transparent or opaque substrate surface and using the resulting product, in sheet form, with or without the substrate, for various decorative or security items.

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In its preferred application, the decorative sheet of the present invention would be laminated via a pressure sensitive adhesive or conventional laminating adhesive to the face of paper board or plastic identification cards, or the like, to complicate the forgery or unauthorized alteration of the cards. In one embodiment, the thickness of the ink layer is small enough so that the card information is visible through the decorative film. In another embodiment, only a portion of the card is covered with an opaque layer which displays an optical decoration, such as a holographic image. Normally, the coated embossed side would be between the sheet and its substrate.

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It is obvious that minor changes may be made in the form and construction of the invention without departing from the material spirit thereof. It is not, however, desired to confine the invention to the exact form herein shown and described, but it is desired to include all such as properly come within the scope claimed.

(11)

The invention having been thus described, what is claimed as new and desire to secure by Letters Patent is:

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### **CLAIMS**

- 1. A method of forming a decorative sheet, comprising the steps of:
- a) forming a sheet of base material, said sheet having a first surface and a second surface,
- b) preparing an ink which is adapted to bond to the first surface of the sheet and to form, with the first surface, an interface which approximates the light reflective properties of an interface formed by vacuum metallization, and
- c) applying the ink to at least a portion of the first surface of the sheet using conventional printing equipment.
  - 2. A method as recited in Claim 1, wherein the first surface is embossed.
- 3. A method as recited in claim 1, wherein the first surface is embossed with an optical diffraction pattern.
- 4. A method as recited in claim 1, wherein the first surface is embossed with a holographic image.
- 5. A method as recited in Claim 1, wherein the first surface is embossed with a geometrical pattern.
  - 6. A method as recited in Claim 1, wherein the sheet is light transmitting.
  - 7. A method as recited in Claim 1, wherein the first surface of the product of step c) is bonded to a backing sheet.
- 8. A method as recited in claim 1, wherein the first surface of the product of step c) is printed with conventional inks.

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- 9. A method as recited in Claim 1, wherein the first surface of the product of step c) is bonded to a backing sheet bearing visual information.
- 10. A method as recited in Claim 9, wherein a pressure sensitive adhesive is used to bond said surface of the product of step c) to said backing sheet bearing visual information.
- 11. A method as recited in Claim 2, wherein the ink is formed of a binder and metallic flakes.
- 12. A method as recited in Claim 11, wherein the ratio of binder to metallic flakes is great enough to bind the flakes to the first surface, and small enough to avoid quenching the optical effect caused by the embossment.
- 13. A method as recited in Claim 11, wherein the volume ratio of binder to metallic flakes is in the range of 0.01 to 2.5.
- 14. A method as recited in Claim 11, wherein the volume ratio of binder to metallic flakes is in the range of 0.5 to 1.0.
- 15. A method as recited in Claim 11, wherein the ink contains sufficient binder to bond the flakes to the first surface while the amount of binder is below the amount required for the binder to substantially coat the first surface with binder.
- 16. A method as recited in Claim 11, wherein the first surface is embossed with an embossment, and the ink contain sufficient binder to bond the flakes to the first surface while the amount of binder is below the amount required for the binder to substantially fill the embossment and quench the optical effect of the embossment.

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- 17. A method as recited in Claim 11, wherein the metallic flakes have a thickness of 100 to 500 Angstroms.
- 18. A method as recited in Claim 11, wherein the metallic flakes have an average diameter of 10 to 20 microns.
- 19. A method as recited in Claim 11, wherein the metallic flakes are aluminum.
- 20. A method as recited in Claim 11, wherein the metallic flakes are formed by vacuum metallizing a thin layer of metal on a carrier.
- 21. A method as recited in Claim 11, wherein the metallic flakes are formed by vacuum metallizing a thin layer of metal on a release layer on a carrier.
- 22. A method as recited in Claim 2, wherein the ink is formed of a crystalline material which possesses metal-like high reflectivity at high concentration.
- 23. A method as recited in Claim 2, wherein the ink is formed of a dye selected from the group consisting of crystal violet, rhodamine B, fuchsine, methyl violet, safranine, fluorescein, rhodamine, and mauveine.
- 24. A method as recited in Claim 2, wherein the ink is formed of a dye selected from the group consisting of crystal violet, and rhodamine B.
  - 25. A method as recited in Claim 2, wherein the ink includes graphite.
- 26. A method as recited in Claim 2, wherein the ink includes the sulfur compounds of elements selected from the group consisting of iron, lead, cobalt, nickel, tin, and zirconium.

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- 27. A method as recited in Claim 2, wherein the ink includes a pigment formed of optical stacks.
- 28. A method as recited in Claim 2, wherein the ink includes a pigment formed of optical stacks having an embossed surface.
- 29. A method as recited in Claim 2, wherein an optical stack is formed on one side of the sheet.
- 30. A method as recited in Claim 1, wherein the ink is formed of a binder and embossed metal flakes.
- 31. A method as recited in Claim 30, wherein the metallic flakes have a thickness of 100 to 500 Angstroms.
- 32. A method as recited in Claim 30, wherein the metallic flakes have an average diameter of 10 to 50 microns.
- 33. A method as recited in Claim 30, wherein the metallic flakes are aluminum.
- 34. A method as recited in Claim 30, wherein the metallic flakes are formed by vacuum metallizing a thin layer of metal on an embossed carrier.
- 35. A method as recited in Claim 30, wherein the metallic flakes are formed by vacuum metallizing a thin layer of metal on an embossed release layer on a carrier.
  - 36. A method as recited in Claim 1, wherein the ink includes graphite.

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- 37. A method as recited in Claim 1, wherein the ink includes the sulfur compounds of elements selected from the group consisting of iron, lead, cobalt, nickel, tin, and zirconium.
- 38. A method as recited in Claim 1, wherein the ink includes a pigment formed of optical stacks.
- 39. A method as recited in Claim 1, wherein the ink includes a pigment formed of optical stacks having an embossed surface.
- 40. A method as recited in Claim 1, wherein an optical stack is formed on one side of the sheet.
- 10 41. A decorative sheet, comprised of:
  - a) a sheet of base material, said sheet having a first surface and a second surface, and b) a layer of ink which is bonded to at least a portion of the first surface of the sheet to form, with the first surface, an interface which approximates the light reflective properties of an interface formed by vacuum metallization.
    - 42. A sheet as recited in Claim 41, wherein the first side is embossed.
  - 43. A sheet as recited in claim 41, wherein the first surface is embossed with an optical diffraction pattern.
  - 44. A sheet as recited in Claim 41, wherein the first surface is embossed with a holographic image.
  - 45. A sheet as recited in Claim 41, wherein the first surface is embossed with a geometrical pattern.

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- 46. A sheet as recited in Claim 41, wherein the sheet is light transmitting.
- 47. A sheet as recited in Claim 41, wherein the first surface of the product is bonded to a backing sheet.
- 48. A sheet as recited in Claim 41, wherein the first surface of the product is additionally printed with conventional inks.
- 49. A sheet as recited in Claim 41, wherein the first surface of the product is bonded to a backing sheet bearing visual information.
- 50. A sheet as recited in Claim 41, wherein a layer of pressure sensitive adhesive is positioned between said first surface and said backing sheet bearing visual information.
- 51. A sheet as recited in Claim 42, wherein the ink is formed of a binder and metallic flakes.
- 52. A sheet as recited in Claim 51, wherein the ratio of binder to metallic flakes is great enough to bind the flakes to the first surface, and small enough to avoid quenching the optical effect caused by the embossment.
- 53. A sheet as recited in Claim 51, wherein the volume ratio of binder to metallic flakes is in the range of 0.01 to 2.5.
- 54. A sheet as recited in Claim 51, wherein the volume ratio of binder to metallic flakes is in the range of 0.5 to 1.0.
- 55. A sheet as recited in Claim 51, wherein the ink contains sufficient binder to bond the flakes to the first surface while the amount of binder is below the amount required for the binder to substantially coat the first surface with binder.

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- 56. A sheet as recited in Claim 51, wherein the first surface is embossed with an embossment, and the ink contain sufficient binder to bond the flakes to the first surface while the amount of binder is below the amount required for the binder to substantially fill the embossment and quench the optical effect of the embossment.
- 57. A sheet as recited in Claim 51, wherein the metallic flakes have a thickness of 100 to 500 Angstroms.
- 58. A sheet as recited in Claim 51, wherein the metallic flakes have an average diameter of 10 to 50 microns.
  - 59. A sheet as recited in Claim 51, wherein the metallic flakes are aluminum.
- 60. A sheet as recited in Claim 51, wherein the metallic flakes are formed by vacuum metallizing a thin layer of metal on a carrier.
- 61. A sheet as recited in Claim 51, wherein the metallic flakes are formed by vacuum metallizing a thin layer of metal on a release layer on a carrier.
  - 62. A sheet as recited in Claim 42, wherein the ink is formed of a dye.
- 63. A sheet as recited in Claim 42, wherein the ink is formed of a dye selected from the group consisting of crystal violet, rhodamine B, fuchsine, methyl violet, safranine, fluorescein, rhodamine, and mauveine.
- 64. A sheet as recited in Claim 42, wherein the ink is formed of a dye selected from the group consisting of crystal violet, and rhodamine B.
- 65. A sheet as recited in Claim 42, wherein the ink includes a pigment formed of optical stacks.

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- 66. A sheet as recited in Claim 42, wherein the ink includes a pigment formed of optical stacks having an embossed surface.
- 67. A sheet as recited in Claim 42, wherein an optical stack is formed on one side of the sheet.
- 68. A sheet as recited in Claim 41, wherein the ink is formed of a binder and embossed metal flakes.
- 69. A sheet as recited in Claim 68, wherein the metallic flakes have a thickness of 100 to 500 Angstroms.
- 70. A sheet as recited in Claim 68, wherein the metallic flakes have an average diameter of 10 to 20 microns.
  - 71. A sheet as recited in Claim 68, wherein the metallic flakes are aluminum.
- 72. A sheet as recited in Claim 68, wherein the metallic flakes are formed by vacuum metallizing a thin layer of metal on an embossed carrier.
- 73. A sheet as recited in Claim 68, wherein the metallic flakes are formed by vacuum metallizing a thin layer of metal on an embossed release layer on a carrier.
  - 74. A sheet as recited in Claim 41, wherein the ink is formed of graphite.
- 75. A sheet as recited in Claim 41, wherein the ink includes a crystalline pigment which has optically metal-like surfaces.
- 76. A sheet as recited in Claim 41, wherein the ink includes a pigment formed of optical stacks.
- 77. A sheet as recited in Claim 41, wherein the ink includes a pigment formed of optical stacks having an embossed surface.

(20)

78. A sheet as recited in Claim 41, wherein an optical stack is formed on one side of the sheet.

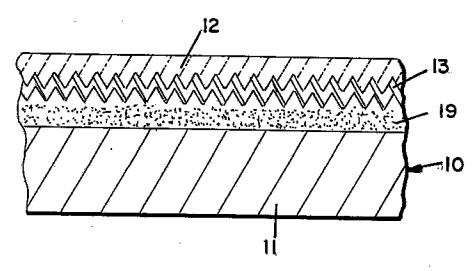


Fig.1

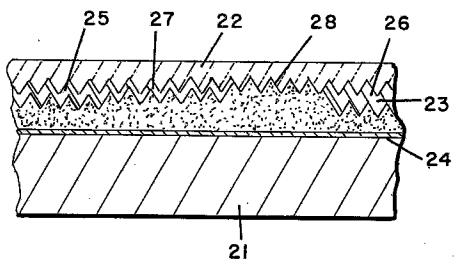
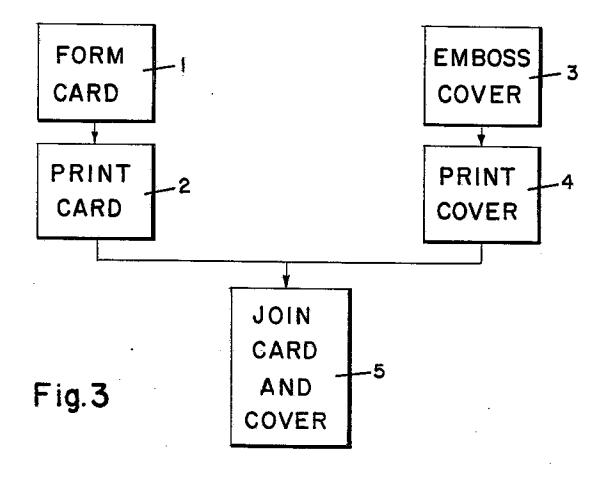
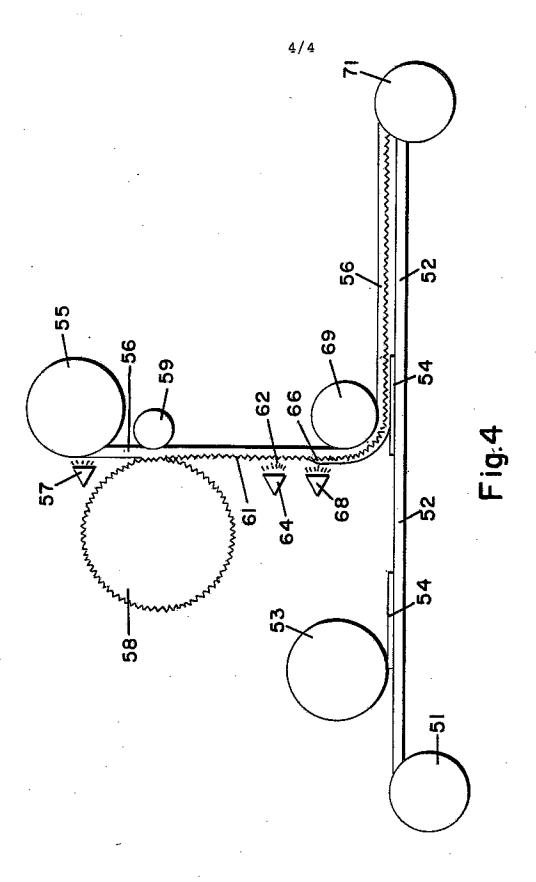


Fig. 2





International Application No

PCT/US 93/04411

I. CLASSIFICATION OF SUBJECT MATTER (if several classification symbols apply, indicate all) <sup>6</sup>						
_	to International Patent . 5 B41M3/14	Classification (IPC) or to both National; B42D15/10;		B44F7/00		
II. FIELDS	SEARCHED		· · · · · · · · · · · · · · · · · · ·			
		Minimum Docu	mentation Searched?			
Classificati	on System		Classification Symbols			
Int.Cl.	. 5	B44C ; B41M ; B44F	B42D ; G03H			
			ner than Minimum Documentation its are included in the Fields Searched <sup>®</sup>			
III. DOCUM	MENTS CONSIDERE	ED TO BE RELEVANT <sup>9</sup>				
Category °	Citation of D	ocument, <sup>11</sup> with indication, where appro	priate, of the relevant passages <sup>12</sup>	Relevant to Claim No. 13		
х	21 Nove	231 572 (THE DE LA RUI mber 1990 e 2, line 1 - page 10		1-3		
<b>X</b>	WO,A,9 22 Marc see pag	1-3				
X A	US,A,5 28 Janu cited i see col	1,37,38, 40,41, 76,78 11,13, 14,18,				
				21,22, 26-30, 32-35		
		<del></del>	<b>-/</b>			
"A" do con fill "L" doc with citt "G" do où "P" doc	nsidered to be of particilier document hat publing date cament which may threigh is cited to establish ation or other special a cument referring to an her means	meral state of the art which is not cular relevance lished on or after the international ow doubts on priority claim(s) or in the publication date of another reason (as specified) a oral disclosure, use, exhibition or r to the international filing date but	"I" inter document published after the it or priority date and not in conflict vected to understand the principle or invention.  "X" document of particular relevance; the cannot be considered novel or cannot involve an inventive step.  "Y" document of particular relevance; the cannot be considered to involve an document is combined with one or ments, such combination being obvious the art.  "A" document member of the same pate.	rith the application but theory underlying the se claimed invention at be considered to se claimed invention avention step when the sore other such docu- ous to a person skilled		
IV. CERT	IFICATION					
Date of the Actual Completion of the International Search  13 SEPTEMBER 1993  23. 09. 93						
internation	al Searching Authority EUROPI	ZAN PATENT OFFICE	Signature of Authorized Officer DOOLAN G.J.			

III. DOCUME	INTS CONSIDERED TO BE RELEVANT (CONTINUED FROM THE SECOND SHEET)					
Category D						
Category	Charlest of Doctment, with managinal, where appropriate, or the research passages	Reevant to Claim No.				
<b>(</b>	EP,A,O 253 543 (THOMAS DE LA RUE & COMPANY) 20 January 1988 see page 1, line 1 - page 3, line 40; example 1	1				
	WO,A,9 004 812 (AUGUST DE FAZIO ET. AL.) 3 May 1990  see page 1, paragraph 1 - page 8, paragraph 2	1,2,4, 11, 18-21, 41,42, 44,51, 58-60				
	EP,A,O 320 638 (MANNINGTON MILLS INC.) 21 June 1989  see page 5, line 35 - page 9, line 47	41,42, 45, 47-49, 51,58,59				

## ANNEX TO THE INTERNATIONAL SEARCH REPORT ON INTERNATIONAL PATENT APPLICATION NO.

US 9304411 SA 74278

This namex lists the patent family members relating to the patent documents cited in the above-mentioned international search report.

The members are as contained in the European Patent Office EDP file on

The European Patent Office is in no way liable for these particulars which are merely given for the purpose of information. 13/09/93

Patent document cited in search report	Publication date	Patent family member(s)		Publication date
GB-A-2231572	21-11-90	None		
W0-A-9002658	22-03-90	EP-A- GB-A- US-A-	0433330 2241668 5199744	26-06-91 11-09-91 06-04-93
US-A-5084351	28-01-92	US-A- US-A- US-A- US-A-	5059245 4434010 5135812 5171363	22-10-91 28-02-84 04-08-92 15-12-92
EP-A-0253543	20-01-88	None		
WO-A-9004812	03-05-90	US-A- CA-A- EP-A- JP-T-	5189531 2000532 0401319 3503811	23-02-93 17-04-90 12-12-90 22-08-91
EP-A-0320638	21-06-89	US-A- AU-A- CA-A-	4863782 2565888 1317456	05-09-89 18-05-89 11-05-93